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Abstract

REPROCESSING OF NUCLEAR FUELS BY VOLATILIZATION

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Since the last Geneva Conference, the technique of volatilization reprocessing has changed: in many cases it has called for gas-solid reactions. Besides, the types of fuel to be taken into consideration are no longer the same : the regeneration of elements based on naturel uranium is not envisaged any more, whereas the ceramics are the object of intensive research.

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The present document forms part of the contribution made by Belgium in this field. Different reprocessing cycles have been studied for some ceramic fuels in the form of oxide or carbide, and for some uranium alloys.

The oxides and the carbides are submitted to gas-solid reactions which, in one way, pulverize the fuel and facilitate the formation of plutonium and uranium hexafluorides, and, in the other way, permit the spreading of the heat liberated during these transformations.

In that way the sequences of the following reactions have been studied:

(1) $UO_2\ (PuO_2) \xrightarrow{O_2} U_3O_8\ (PuO_2) \xrightarrow{NH_3} UO_2(PuO_2) \longrightarrow UF_4(PuF_4) \xrightarrow{F_2} UF_6(PuF_6) \xrightarrow[\text{salts}]{\text{fused}} UO_2(PuO_2)$

(2) $UO_2\ (PuO_2) \xrightarrow{O_2} U_3O_8(PuO_2) \xrightarrow{HF} UF_4\text{-}UO_2F_2(PuF_4) \xrightarrow{F_2} UF_6(PuF_6) \xrightarrow[\text{salts}]{\text{fused}} UO_2(PuO_2)$

(3) $UO_2(PuO_2) \xrightarrow{O_2} U_3O_8(PuO_2) \xrightarrow{HF} UF_4\text{-}UO_2F_2(PuF_4) \xrightarrow{ClF_3} UF_6(PuF_4) \xrightarrow{F_2} (PuF_6) \xrightarrow[\text{salts}]{\text{fused}} (PuO_2)$

(4) $UO_2\ (PuO_2) \xrightarrow[O_2]{HNO_3} UO_3\ (PuO_2) \xrightarrow{SF_4} UF_6(PuF_4) \xrightarrow{F_2} (PuF_6) \xrightarrow[\text{salts}]{\text{fused}} (PuO_2)$

(5) $UC\ (PuC) \xrightarrow{HF} UF_4(PuF_4) \xrightarrow{ClF_3} UF_6(PuF_4) \xrightarrow{F_2} (PuF_6) \xrightarrow[\text{salts}]{\text{fused}} (PuO_2)$

(6) $UO_2 \xrightarrow[Cl_2]{CCl_4} UCl_6 \xrightarrow[H_2]{H_2O} U\ O_2$

Some of these cycles have been combined among themselves. The first and the fourth have been discarded, because of technological difficulties, as well as the long time required for the reactions, and their low yield. Preference has been given to cycles (2) and (3). These are characterized by the use of fluorine to effect the simultaneous volatilization of the uranium and the plutonium, or of chlorine trifluoride to obtain a successive volatilization of these two compounds; this successive volatilization appears particularly advantageous, because it permits the plutonium hexafluoride to be treated in an apparatus of small volume.

Although no operations have been performed on real fuels,

the results of pilot studies on cycles (2) and (3) give reason to believe that the recovery of uranium will surpass 99 per cent. A large technological effort has still to be made to apply the basic knowledge already acquired for the separation of uranium and plutonium and for the recuperation of the latter, in a suitable form.

Cycle (5) proposed for the reprocessing of carbides is a particularly interesting example of the great simplicity which the volatilization methods can attain.

The last cycle, schematised in the sequence of reactions (6), has stimulated an extensive study of the synthesis and properties of uranium chlorides and of those of fission products, as well as their complexes. Up to now it has not received concrete realizations.

The study of processing of alloys has pertained to the following elements: U-Zr, U-Mo, U-Nb, U-Al. Some chemical conditions of reaction and separation under the action of chlorinating agents have been studied. For the uranium-aluminium alloy, the research has evolved towards a cycle of processing by chlorination and fluorination, where hydrochloric acid is called upon to volatilize the aluminium; the residual uranium trichloride can subsequently be transformed into hexafluoride.

In the scope of the European economy, the processes proposed will be the most paying for fuels rich in fissile materials. Thus, volatilization should rather be considered as complementary to liquid-liquid extraction, of which the actual principal applications concern low enrichments. The numerous unsolved technological problems, as well as the necessity to verify the data acquired by preliminary work, cause a delay of 5 to 10 years to be foreseen before the birth of the first industrial applications.